Ultrasharp Lines in the Absorption and Fluorescence Spectra of an Atom in a Cavity

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The absorption and fluorescence spectra of an atom driven by a strong, in general, off-resonance external field and coupled to a single cavity mode is investigated. It is shown that for moderate coupling constant the spectral components can become ultrasharp lines whose linewidths are narrower than both the natural linewidth and the cavity decay rate.

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Sharp lines in absorption and fluorescence are important for precision atomic spectroscopy [1,2] and are the subject of intense studies [1–5]. One well-established method of producing sharp spectral lines is that of saturation spectroscopy, where a weak probe beam is used to study a medium saturated by an intense driving field (or "saturator wave"). In media dominated by dephasing collisions, a "coherent dip" appears in plots of absorption versus probe detuning [1,2]. Such dips allow one to measure population decay times in liquids or semiconductors. The linewidths of absorption and fluorescence spectra can also be suppressed by a squeezed reservoir [3,4]. The effect of a cavity on the linewidth in spontaneous emission and resonance fluorescence spectra has been discussed in Refs. [5–10]. However, so far, only the strong coupling limit has been considered, and in this limit only 50% linewidth narrowing can be reached [5]. Obviously, strong coupling is the most difficult experimental condition to achieve in the optical domain [8,9].

In this Letter, we study the saturation spectroscopy of an atom in a cavity. We obtain the interesting result that for moderate coupling constant, the probe absorption spectrum displays one absorptive and one amplifying resonance at the Mollow sideband frequencies, whose linewidths are each much smaller than both the natural linewidth and the cavity decay rate. Furthermore, the resonance fluorescence spectrum of the strongly driven atom (corresponding to the same system parameters) also contains three ultrasharp lines at the Mollow triplet frequencies. We emphasize that these sharp fluorescence lines result from inelastic, not elastic, scattering.

Our system consists of a two-level atom with excited state $|2\rangle$, ground state $|1\rangle$, and transition frequency ω_a. The atom is driven by a coherent external field at a frequency ω_L with resonant Rabi frequency 2ϵ, coupled to a cavity mode with coupling constant g, and damped at the rate γ by spontaneous emission to modes other than the privileged cavity mode. The cavity mode frequency ω_c is chosen so that $\omega_c=\omega_L+2\Omega$, where $2\Omega=2\epsilon\sqrt{1+\delta^2}$ is the detuned Rabi frequency and $\delta=(\omega_a-\omega_L)/2\epsilon$ is the scaled detuning; the cavity decay rate is k. The behavior of the system is governed by the master equation for the density operator ρ [5,6,10,11]

$$\frac{\partial}{\partial t}\rho=-i[H,\rho]+\frac{1}{2}\gamma L_a\rho+\frac{1}{2}kL_c\rho\,, \tag{1}$$

where

$$H=H_{da}+H_c+W\,, \tag{2}$$

$$L_a\rho=2\sigma_{12}\rho\sigma_{21}-\sigma_{21}\sigma_{12}\rho-\rho\sigma_{21}\sigma_{12}\,, \tag{3}$$

and

$$L_c\rho=2a\rho a^\dagger-a^\dagger a\rho-\rho a^\dagger a\,. \tag{4}$$

Here $L_a\rho$ and $L_c\rho$ are operators, representing the damping of the atom by spontaneous emission and of the cavity field by cavity decay; σ_{ij} are the atomic operators $\sigma_{ij}=|i\rangle\langle j|$ $(i,j=1,2)$, and a and $a^\dagger$ are the cavity-mode (boson) annihilation and creation operators; H_{da} is the Hamiltonian of the dressed atom (atom plus driving field), and has eigenstates $|\tilde{i},N\rangle$ $(\tilde{i}=\tilde{1},\tilde{2})$ satisfying the eigenvalue equation [12]

$$H_{da}|\tilde{i},N\rangle=[N\omega_L+(-1)^i\Omega]|\tilde{i},N\rangle\,, \tag{5}$$

where

$$|\tilde{1},N\rangle=\cos\phi|1,N\rangle-\sin\phi|2,N-1\rangle\,, \tag{6}$$

$$|\tilde{2},N\rangle=\sin\phi|1,N\rangle+\cos\phi|2,N-1\rangle\,, \tag{7}$$

$|i,N\rangle$ is the state in which the atom is in state $|i\rangle$ and N photons are present in the driving mode, and ϕ is given by

$$\cos^2\phi=\frac{1}{2}+\frac{\delta}{2\sqrt{1+\delta^2}}\,. \tag{8}$$

H_c is the Hamiltonian of the cavity mode, and has the eigenvalue equation

$$H_c|n\rangle=n(\omega_L+2\Omega)|n\rangle\,. \tag{9}$$

The noninteracting dressed-atom+cavity-mode Hamiltonian $H_0=H_{da}+H_c$ has the eigenvalue equation

$$H_0|\tilde{i},N,n\rangle=[N\omega_L+(-1)^i\Omega+n(\omega_L+2\Omega)]|\tilde{i},N,n\rangle\,, \tag{10}$$

where $|\tilde{i},N,n\rangle=|\tilde{i},N\rangle\otimes|n\rangle$. The state $|\tilde{i},N,0\rangle$ is nondegenerate, while the states $|\tilde{2},N,n\rangle$ and $|\tilde{1},N-1,n+1\rangle$ form doubly degenerate pairs. When we include the in-

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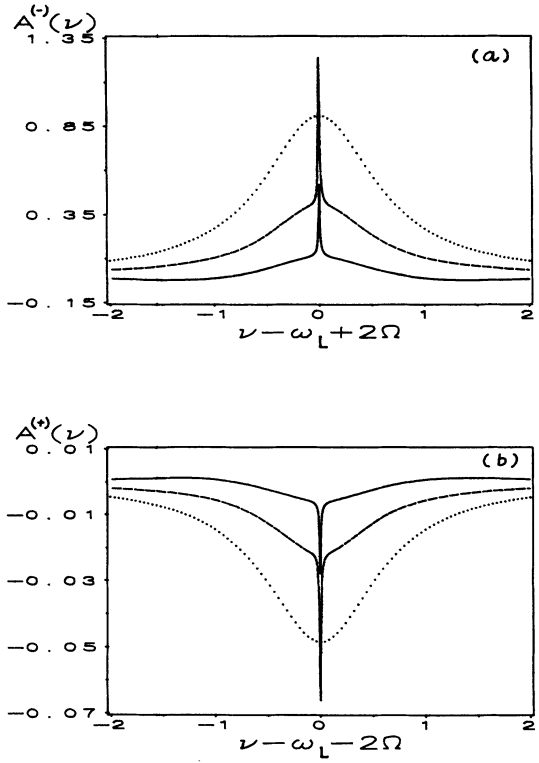

FIG. 1. Absorption spectrum (in units of $q=1$, $\gamma=1$) $A^{(-)}(\nu)$ (a) and $A^{(+)}(\nu)$ (b) for $k=0.05$, $\delta=-0.8$, and for $g=0.01$ (dotted curves), $g=0.8$ (dashed curves), and $g=1.25$ (solid curves).

teraction W between the dressed atom and cavity mode, $W=g(a^{\dagger}\sigma_{12}+\sigma_{21}a)$, the degeneracy is lifted, resulting in doublets $|N,\pm n\rangle$ which satisfy the eigenvalue equation

$$(H_0+W)|N,\pm n\rangle=E_{N,\pm n}|N,\pm n\rangle\,, \tag{11}$$

where

$$E_{N,\pm n}=N\omega_L+(2n-1)\Omega\pm g_1\sqrt{n}\,, \tag{12}$$

$$|N,\pm n\rangle=\frac{1}{\sqrt{2}}(|\tilde{2},N-n+1,n-1\rangle\pm|\tilde{1},N-n,n\rangle)\,, \tag{13}$$

for $n\neq 0$, $|N,0\rangle=|\tilde{1},N,0\rangle$, and $g_1=g\cos^2\phi$.

The fluorescence spectrum of this system was calculated in Ref. [10] for the strong coupling limit, $g\gg\gamma,k$, both by a numerical, vector continued-fraction solution of the master equation and by an analytical solution, using the doubly dressed-atom states described above. For details of both solutions, we refer the reader to Ref. [10]. In this Letter, we focus on the case of moderate coupling, which we have found to produce dramatically different spectra, and on the absorption spectrum of a weak probe beam by the system.

The absorption spectrum has the well-known form [13]

$$A(\nu)=q\mathrm{Re}\int_0^{\infty}e^{i\nu\tau}\langle[\sigma_{12}(\tau),\sigma_{21}]\rangle_s\,d\tau\,, \tag{14}$$

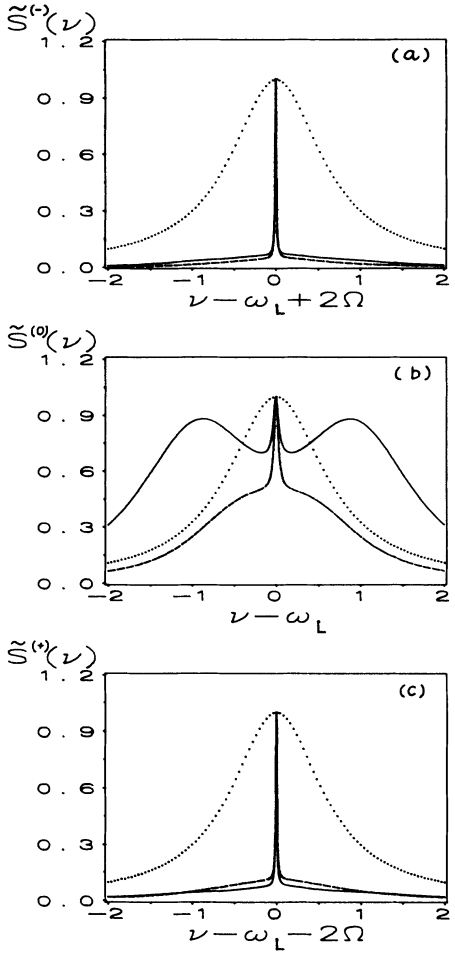

FIG. 2. Inelastic normalized fluorescence spectrum $\tilde{S}^{(-)}(\nu)=S^{(-)}(\nu)/S^{(-)}(\omega_L-2\Omega)$ (a), $\tilde{S}^{(0)}(\nu)=S^{(0)}(\nu)/S^{(0)}(\omega_L)$ (b), and $\tilde{S}^{(+)}(\nu)=S^{(+)}(\nu)/S^{(+)}(\omega_L+2\Omega)$ (c) for the same parameters as in Fig. 1.

where s indicates an average over the steady-state solution of the master equation, and q is a normalization constant containing geometric and atomic factors. For high Ω, $A(\nu)$ consists of two well-separated sidebands, centered at the frequencies $\omega_L\pm 2\Omega$. This spectrum is calculated numerically by the vector continued-fraction techniques described in Refs. [10,14] and plotted in Fig. 1 for small and moderate values of the coupling constant g. For small coupling, $g=0.01\gamma$ (dotted curves), the absorption spectrum is almost the same as in free space. The minus sideband $A^{(-)}(\nu)$ [Fig. 1(a)] is positive, corresponding to a net absorption by the system, and the plus sideband $A^{(+)}(\nu)$ [Fig. 1(b)] negative, corresponding to a net amplification of the probe beam. With increasing g, $A^{(\pm)}(\nu)$ become very narrow peaks: For $g=0.8\gamma$ (dashed lines) the width is about 0.015γ, and for $g=1.25\gamma$ (solid lines) 0.01γ, much narrower than both γ and the cavity decay rate $k=0.05\gamma$.

In Fig. 2 we plot the inelastic part of the normalized fluorescence spectrum, calculated as in Ref. [10], for the

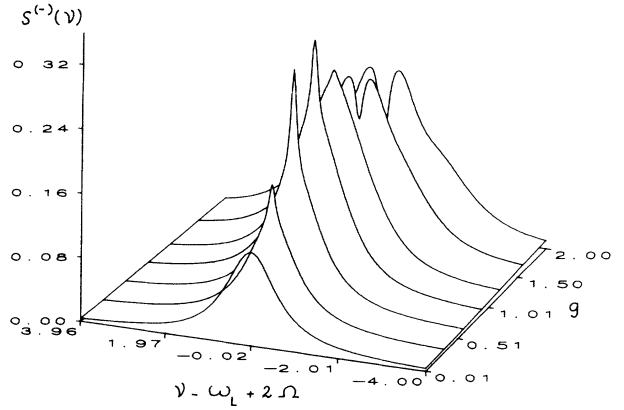

FIG. 3. Left sideband $S^{(-)}(\nu)$ of fluorescence spectrum (in units of $\gamma=1$) for $\delta=-0.4$, $k=0.2$, and for various values of g.

same system parameters as in Fig. 1. Like the absorption spectrum, the fluorescence spectrum develops ultrasharp lines for moderate g, particularly at the sideband frequencies $\omega_L\pm2\Omega$.

Finally, in Fig. 3 we plot a three-dimensional picture showing the development of the fluorescence sideband $S^{(+)}(\nu)$ with increasing g from its (almost) free-space shape for small g ($g=0.01\gamma$), through its sharp-line shape for moderate g ($g\sim\gamma$), and beginning to split into a doublet (Ref. [10]) for higher coupling. In this figure we have deliberately chosen a (relatively) large value of k ($k=0.2\gamma$) in order to lower the height of the sharp line so that it can be displayed in the same figure with the other spectra.

The region of parameter space for which this unusual behavior occurs involves only moderate coupling ($g_1<\gamma$), but at the same time (at least) moderately high $\langle n\rangle$. This combination is easily achieved by negative detuning of the laser (Ref. [11]). The explanation for its behavior can be obtained from the dressed-atom picture. As discussed in detail in Ref. [10], the sidebands $A^{(+)}(\nu)$ and $S^{(+)}(\nu)$ are due to transitions of the doubly dressed atom between states $|N,\pm(n+1)\rangle$ and $|N-1,\pm n\rangle$, giving rise to spectral lines at frequencies $\omega_L+2\Omega\pm\nu_n^{(\pm)}$,

$$\nu_n^{(\pm)}=g_1(\sqrt{n+1}\pm\sqrt{n})\,. \tag{15}$$

Similarly, the sidebands $A^{(-)}(\nu)$ and $S^{(-)}(\nu)$ involve radiative transitions between the states $|N,\pm n\rangle$ and $|N-1,\pm(n+1)\rangle$, giving rise to lines at frequencies $\omega_L-2\Omega\pm\nu_n^{(\pm)}$. [There is a central component $S^{(0)}(\nu)$ in the fluorescence spectrum, resulting from transitions from $|N,\pm n\rangle$ to $|N-1,\mp n\rangle$ at frequencies $\omega_L\pm2g_1\sqrt{n}$. However, the central component in the absorption spectrum vanishes, just as it does in free space, due to cancellation between absorption and stimulated emission processes.] For large values of g, both the $\nu_n^{(+)}$ and $\nu_n^{(-)}$ lines in the sidebands are well resolved, producing multiplet spectra as shown for $S^{(\pm)}(\nu)$ in Ref. [10]. For moderate g, the $\nu_n^{(+)}$ lines can still be treated as resolved, producing broad features in the spectra with peaks centered at $\sim\omega_L\pm2g_1\sqrt{\langle n\rangle}$ and at $\omega_L\pm2\Omega\pm2g_1\times\sqrt{\langle n\rangle}$ where $\langle n\rangle$ is the mean photon number in the cavity mode. The $\nu_n^{(-)}$ (sideband) lines, however, are *not* resolved for moderate g and higher n:

$$\nu_n^{(-)}\Rightarrow\frac{g_1}{2\sqrt{n}}\ll\gamma\,. \tag{16}$$

In calculating the spectrum, we therefore cannot neglect the coupling by spontaneous emission of the off-diagonal elements of ρ within the different sets of frequencies $\omega_L+2\Omega\pm\nu_n^{(-)}$ and $\omega_L-2\Omega\pm\nu_n^{(-)}$. We have calculated the spectrum of the absorption and fluorescence sidebands by the dressed-atom model, including the coherence among these different matrix elements of ρ, and have reproduced analytically the ultrasharp line results. We outline here briefly the calculation corresponding to the plus sidebands $A^{(+)}(\nu)$ and $S^{(+)}(\nu)$, which involve the evolution of the (averaged over N) density matrix elements $\rho^{\pm}_{\pm(n+1),\pm n}=\langle\rho^{\pm}_{\pm(n+1),\pm n,N}\rangle_N$, where $\rho^{+}_{n+1,n,N}=|N,n+1\rangle\langle N-1,n|$.

The evolution of these elements is obtained by projecting the master equation in turn onto $|N-1,\pm n\rangle$ on the right and $\langle N,\pm(n+1)|$ on the left, in all possible combinations, without making any secular approximations in considering the coherences among the $\nu_n^{(-)}$ lines. We define the dipole transition moments $p_{\pm n}$,

$$p_{\pm n}\equiv\mu_{\pm(n+1),\pm n}\rho^{(+)}_{\pm(n+1),\pm n}\,, \tag{17}$$

where $\mu_{n,n'}=\langle N,n|\sigma_{21}|N-1,n'\rangle$. Equations of motion for the associated quantities $P_n\equiv p_n+p_{-n}$, and $M_n\equiv p_n-p_{-n}$ can be found from master equation (1) in the form

$$\dot{P}_n=-i(\omega_L+2\Omega)P_n-i\nu_n^{(-)}M_n-(C_n+A_n)P_n+A_{n+1}P_{n+1}\,, \tag{18}$$

$$\dot{M}_n=-i(\omega_L+2\Omega)M_n-i\nu_n^{(-)}P_n-(D_n+B_n)M_n+\frac{\gamma}{2}\sin^4\phi(M_{n-1}-M_n)+B_{n+1}M_{n+1}\,, \tag{19}$$

where

$$C_n=\Gamma_+\frac{k}{2}[n-\sqrt{(n+1)(n-1)}]\Rightarrow\Gamma_+\frac{k}{4n}\,, \tag{20}$$

$$D_n=\frac{k}{2}[2n-\sqrt{n(n-1)}-\sqrt{n(n+1)}]\Rightarrow\frac{k}{8n}\,, \tag{21}$$

$$A_n=\frac{k}{2}[n+\sqrt{(n+1)(n-1)}]\,, \tag{22}$$

$$B_n=\frac{\gamma}{2}\cos^4\phi+\frac{k}{2}[\sqrt{(n-1)n}+\sqrt{n(n+1)}]\,. \tag{23}$$

Here $\Gamma_+=(\gamma/2)(1+2\sin^2\phi\cos^2\phi)$ is the half-width of the Mollow sidebands in free space.

Equations (18) and (19) contain terms $\nu_n^{(-)}$, C_n, and D_n which vary slowly with n, and which we can replace

by $\nu_{\langle n\rangle}^{(-)}$, $C_{\langle n\rangle}$, and $D_{\langle n\rangle}$. The spectra, however, involve the Fourier transform of the sum over n of (two-time averages of) the transition dipole moments $p_{\pm n}$. Performing the sum over n in Eqs. (18) and (19) and neglecting "end effects" at $n=0$, we obtain equations for $P=\sum_n P_n e^{i(\omega_L+2\Omega)t}$ and $M=\sum_n M_n e^{i(\omega_L+2\Omega)t}$ in the form

$$\dot{P}=-\left[\Gamma_+ + \frac{k}{4\langle n\rangle}\right]P - i\frac{g_1}{2\sqrt{\langle n\rangle}}M\,, \tag{24}$$

$$\dot{M}=-\frac{k}{8\langle n\rangle}M - i\frac{g_1}{2\sqrt{\langle n\rangle}}P\,. \tag{25}$$

The lowest eigenvalue is easily found to be

$$\lambda^{(0)}\approx\frac{k}{8\langle n\rangle}+\frac{g^2\cos^4\phi}{4\Gamma_+\langle n\rangle}\,, \tag{26}$$

which corresponds to the ultrasharp lines. From Ref. [11] we know that for $k\ll g,\gamma$ and $\cos^2\phi\ll 1$, the mean photon number of the cavity field $\langle n\rangle$ is large. Clearly in this case the linewidth of the absorption and fluorescence spectra at frequencies $\omega_L\pm 2\Omega$ is much smaller than both γ and k. The analytical linewidth (26) is found to be in good agreement with the numerical linewidth for large $\langle n\rangle$.

The situation is somewhat different within the central component of the fluorescence triplet. Here, the coherences "corresponding" to the $\nu_n^{(-)}$ lines are $\rho_{nn}^{(+)}$ and $\rho_{-n,-n}^{(+)}$, which interfere for all values of the system parameters, as they all oscillate at the same frequency ω_L. However, all corresponding dipole moment transition elements μ_{nn} vanish, except for μ_{00} [10]. This has two effects in the spectrum: (i) Although the ρ_{nn}^{+} $(n\neq 0)$ all contribute to the evolution of ρ_{00}^{+}, only $p_0=\mu_{00}\rho_{00}^{+}$ actually appears in the spectrum. No sum over n is possible as was performed for the sidebands, and which led to the very-sharp line narrowing. As a result, the line at ω_L is not as sharp as those at $\omega_L\pm 2\Omega$. (ii) The intensity of this line is proportional to P_0; hence the line vanishes as $\langle n\rangle$ increases.

In conclusion, we have calculated the absorption and fluorescence spectra of a strongly driven atom in a cavity. We have shown that for moderate coupling constant $g\approx\gamma$, the spectral components, in particular the sidebands, can become ultrasharp lines, whose linewidths are much narrower than both the natural linewidth and the cavity decay rate. This condition of moderate coupling should be well within current experimental reach.

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